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High Critical Temperature Superconductor Substrate and Buffer Layer Compounds, A_2MeSbO_6 (Where A=Ba and Sr; and Me=Sc, In and Ga)

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13. ABSTRACT *(Maximum 200 words)*

Compounds in the series A2MeSbO6, where A=Ba, Sr, and Me=Sc, In, and Ga, have been used as substrate buffer layers with YBa2Cu3O7-x thin films. These materials were prepared by solid-state reaction of the oxides and carbonates. The compounds are ordered perovskites except for Ba2InSbO6. All compounds are cubic except Sr2ScSbO6 and Sr2GaSbO6 which are pseudo-cubic, tetragonal. Dielectric constant and loss tangent are reported for each bulk compound. Herein is described the successful deposition of thin films A2MeSbO6 on (100) MgO and A2MeSbO6/(001) YBCO/(100) MgO by pulsed laser ablation.

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High Critical Temperature Superconductor Substrate and Buffer Layer Compounds, A_2MeSbO_6 (Where A=Ba, Sr and Me=Sc, In and Ga)

1. INTRODUCTION

Considerable progress has been achieved in the variety and quality of microwave devices that have been fabricated using superconductors. Because most of these devices are in the form of thin films, they require dielectric substrates and/or buffer layers with very exacting properties [1-4] .The most useful dielectrics are lanthanum-aluminate ($LaAlO_3$), magnesium-oxide (MgO), buffered yttrium-stabilized-zirconia (YSZ) and buffered sapphire, all of which have some drawbacks. We have been investigating new substrates in an attempt to overcome various deficiencies in these dielectrics. Compounds in the series A_2MeSbO_6, where A=Ba, Sr and Me=Sc, In and Ga, have been prepared by solid state reaction between oxides and carbonates. When A=Ba and Me = rare earth, an analogous series results, as previously reported [5]. Yet another series where A=Sr and Me=rare-earth, has been reported [6]. All of the reported Ba compounds are cubic and all of the Sr compounds are tetragonal, except for Me=Lu or La which are cubic. Goldschmidt's tolerance factor [7], $t = (r_{a+} r_0)/\sqrt{2}\ (r_{b+} r_0)$, predicts that $0.8< t < 0.9$ perovskite compounds should be cubic. For the barium rare-earth compounds which are cubic, $0.8\leq t \leq 0.9$. For the Sr rare-earth compounds which are mostly non-cubic, $0.96 \leq t \leq 0.99$ [7]. In the present instance, A_2MeSbO_6, the tolerance factor predicts all the compounds to be cubic.

We have previously reported the successful pulsed-laser deposition (PLD) on thin films of strontium-rare-earth-antimonate (Sr_2ReSbO_6) on yttrium-barium-copper-oxide ($YBa_2Cu_3O_{7-\delta}$ or YBCO), [Sr_2ReSbO_6/(00l)YBCO/(100)MgO] [7]. Similar results have been achieved with barium-rare-earth-antimonate (Ba_2ReSbO_6) on YBCO, [Ba_2ReSbO_6/(00l)YBCO/(100)MgO] [8]. Herein is described the successful deposition of thin film A_2MeSbO_6 on YBCO, A_2MeSbO_6/(00l) YBCO/(100)MgO.

2. EXPERIMENTAL

The A_2MeSbO_6 compounds were prepared by mixing reagent grade strontium-carbonate ($SrCO_3$), barium-carbonate ($BaCO_3$), antimony-trioxide (Sb_2O_3), indium-trioxide (In_2O_3), gallium-trioxide (Ga_2O_3) and scandium-trioxide (Sc_2O_3) in stoichiometric proportions by grinding together in a mortar, pressing the mixtures into discs, heating the discs to 1100°C (rate =180°C/h) for 15 h in air, then cooling to 100°C (same rate) and removing from the furnace. Each disc was ground to a powder (100 μm particle size), pressed into 1 and 1/4 inch discs in a steel die, repressed at 60 000 psi isostatically, sintered between 1300 and 1600°C for 20-50 h, and cooled to 100°C and removed from the furnace .

If x-ray diffractometer scans revealed the presence of a second phase, the discs, were reground and the last step was repeated until a single phase was achieved. When second phases were present they often appeared only on the surface and could be removed by grinding. Diffractometer

scans were taken from 15-155° 2θ using CuKα radiation (λ = 1.5405 Å). Lattice parameters were obtained by least squares fit to the Nelson-Riley function [9].

Density measurements were made on bulk samples by He gas pycnometry and compared with calculated x-ray densities in Table 2. The discs were employed as targets for deposition of the films by pulsed-laser deposition (PLD) technique. Deposition parameters were pulse-repetition rate of 10 Hz, laser fluence of 1-2 J/cm^2 at the target,.and oxygen partial pressure and substrate temperature as given in Table 1.

TABLE 1. CONDITIONS FOR DEPOSITION OF MULTILAYERED STRUCTURES.

SAMPLE No.	COMPOUND	SUBSTRATE		OXYGEN PRESSURE (mTorr)	BLOCK TEMPERATURE (°C)
		MgO	YBCO/MgO		
895	Ba_2InSbO_6	X	X	150	800
894	Ba_2ScSbO_6	X	X	150	800
883	Sr_2ScSbO_6	X	X	1	500
881	Sr_2ScSbO_6	X	X	1	400
880	Sr_2ScSbO_6	X	X	1	300
869	Sr_2ScSbO_6		X	15	550
868	Sr_2ScSbO_6		X	1	500
852	$Sr_2InSbO6$		X	30	600
826	Sr_2GaSbO_6		X	170	800
815	Sr_2ScSbO_6	X	X	170	800
814	Sr_2GaSbO_6	X	X	170	800

Microwave measurements of the real and imaginary parts of the dielectric constant were performed at approximately 9.32 and 10.1 GHz and room temperature. A cavity perturbation technique was used with a reflection-type rectangular cavity excited in either the TE_{106} mode for 9.3 GHz or the TE_{107} mode for 10.1 GHz. The cavity was coupled to the waveguide by an adjustable iris. A 0.5 mm by 35 mm slot cut along the center of one of the broad sides of the cavity provides access for the thin, rectangular samples. The samples were held, such that their long dimension was parallel to the E-field of the cavity, and they were positioned at the E-field maximum, as determined by maximizing the shift of the cavity.

The real part of the dielectric constant is calculated from the shift in the resonance frequency of the cavity due to the sample, and the imaginary component is calculated from a change in the cavity Q. The accuracy of these measurements depends upon two general sources of error:

(1) the accuracy of the cavity characterization; and

(2) the material properties such as density and uniformity of shape.

The error due to the cavity characterization results in an accuracy of ±2% for the real part of the dielectric constant, and limits the resolution of the loss tangent (the imaginary component divided by the real component of the loss tangent) to approximately 0.001. The error due to material properties such as sample shape and material density can be considerably greater than the cavity characterization error. However, most sample densities were near 100% of theoretical density and thus should not introduce a large error.

3. RESULTS

All compounds in the series A_2MeSbO_6, except barium-indium-antimonate (Ba_2InSbO_6), occur as ordered perovskites at room temperature. In an earlier investigation [11], Ba_2InSbO_6 was found to be ordered when prepared at 1100°C using barium-dioxide (BaO_2). Only strontium-indium-antimonate (Sr_2InSbO_6) is cubic; the other Sr compounds are tetragonal (pseudo cubic). X-ray diffraction patterns are shown in Fig.1. Lattice parameters and microwave properties may be found in Table 2.

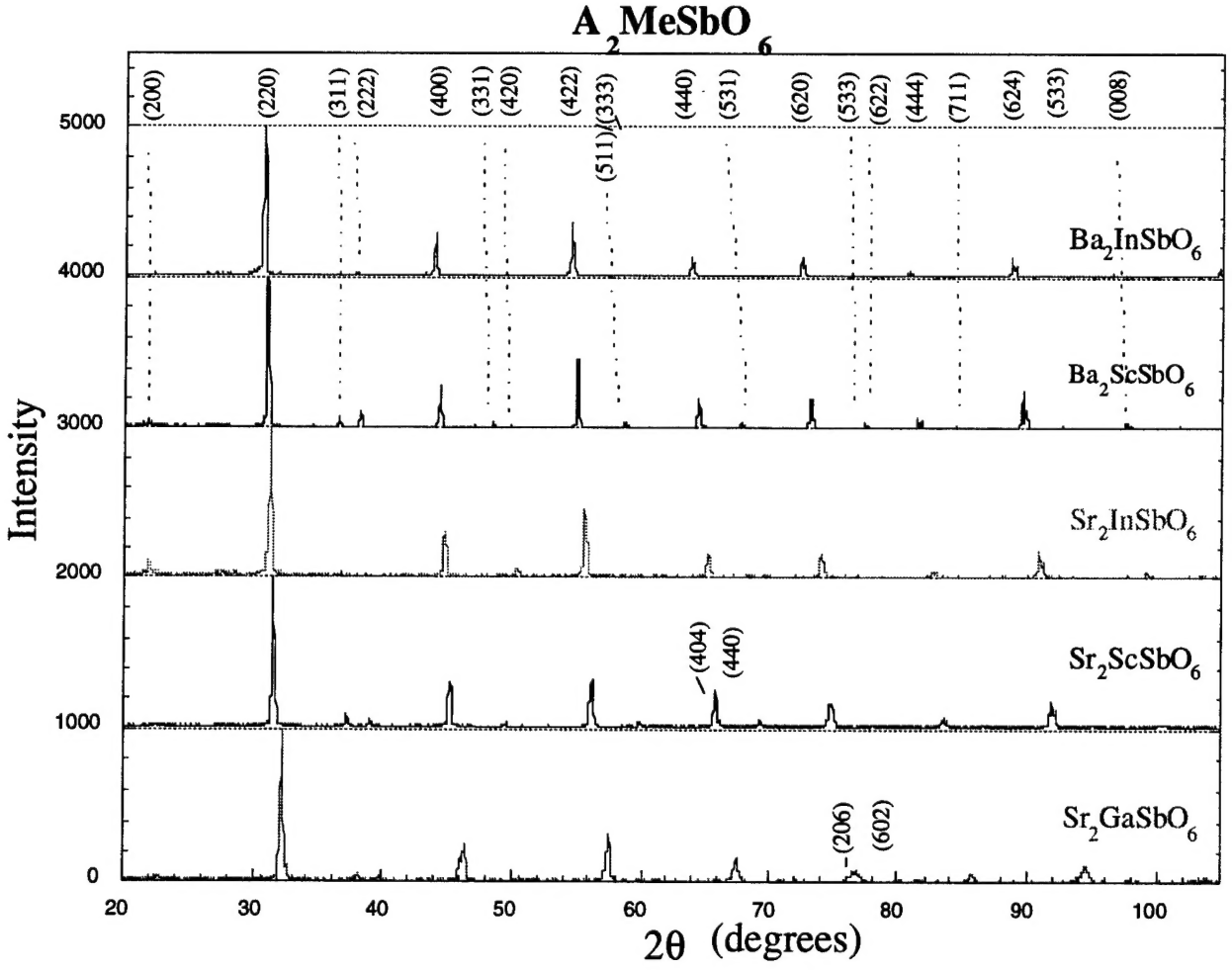

Figure 1. X-ray spectra of A_2MeSbO_6 bulk target samples. The x-ray lines were used to index the lattice parameters, Table 2. The FWHM of the 004 lines of the A_2MeSbO_6 compounds ranged from 0.19-29 2θ.

Cu K_α was used as the radiation source.

TABLE 2. PROPERTIES OF A_2MeSbO_6 COMPOUNDS.

Compound	Lattice Parameter (Å)		Sintering Temp. (°C)	X-Ray Density (g/cm^3)		Dielectric		Color in Bulk
	Literature	Measured		x-ray	exp	Constant	Loss tangent (10^{-3})	
Ba_2InSbO_6	(4.128)[12] (4.136)[13] (8.269)[11]	a=4.174 a=4.134	1300 1400	7.33	7.31	6.9	20	Black
Ba_2ScSbO_6	(8.203)[12] (4.096)[13] (8.197)[11]	a=8.172 a=8.196	1400 1600	6.54	6.59	8.1	6.5	White
Sr_2InSbO_6	(8.095)[12] (4.051)[13]	a=8.094 a=8.096 a=8.086	1200 1400 1600	6.34	6.30	8.3	<0.1	White
Sr_2ScSbO_6	(a=5.678)[12] (b=5.691)[12] (c=8.021)[12] (4.011)[13]	a=8.019 c=8.063	1400	5.49	5.08	8.8	2.1	White
Sr_2GaSbO_6	(a=7.84)[11] (c=7.91)[11] (7.892)[12] (3.924)[13]	a=7.860 c=7.910 a=7.880 c=7.784	1400 1500	6.22	6.12	13.5	9.5	White

When the A_2MeSbO_6 compound which formed exhibits an ordered cubic perovskite structure, a subcell and a supercell were sometimes observed. The most probable space group in this case is Fm3m. The lattice constant frequently contracted as the sintering temperature increased. For the two tetragonal Sr compounds, strontium-scandium-antimonate (Sr_2ScSbO_6) and strontium-gallium-antimonate (Sr_2GaSbO_6), splitting of the pseudo-cubic peaks is visible above $2\theta > 65°$ (Fig.1).

$LaAlO_3$ and MgO are often used as substrates on which high critical temperature superconducting films, such as YBCO, are grown for device applications. The lattice mismatch for heteroepitaxial growth of YBCO on $LaAlO_3$ is outstanding, about 1%; for MgO it is about 7%. The mismatch in lattice parameter for A_2MeSbO_6 compounds epitaxially grown on YBCO is given in Table 3.

Table 3. PERCENT MISFIT TO YBCO

Compound	*a* Lattice Parameter (Å)	% misfit to YBCO	
		a-axis	*b*-axis
Ba_2InSbO_6	a=4.174	8.2	6.2
Ba_2ScSbO_6	a=8.196	7.2	5.3
Sr_2InSbO_6	a=8.086	5.8	3.9
Sr_2ScSbO_6	a=8.019	4.9	3.0
Sr_2GaSbO_6	a=7.880	3.1	1.3

With regard to % misfit, only one compound, Sr_2GaSbO_6, compares favorably with $LaAlO_3$; all but Ba_2ScSbO_6 are superior to MgO. In addition, the compounds of the series contain no magnetic ions and should exhibit no magnetic loss at any temperature.

Analysis of x-ray diffraction data obtained from single layer films of A_2MeSbO_6 on (100) MgO reveals that the 400 reflection of the A_2MeSbO_6 perovskite phase, when ordered, or 200 when not, is the most intense(see Table 4). The high quality of the A_2MeSbO_6 layer on YBCO (001) / (100) MgO is exhibited not only by the sharpness of x-ray diffraction lines (FWHM ≈ 0.30 2θ for 004 reflection) but also by the presence of high-order reflection, 008(see Fig. 2). Crystallization of these thin-film A_2MeSbO_6 materials is observed at deposition temperatures as low as 600°C.

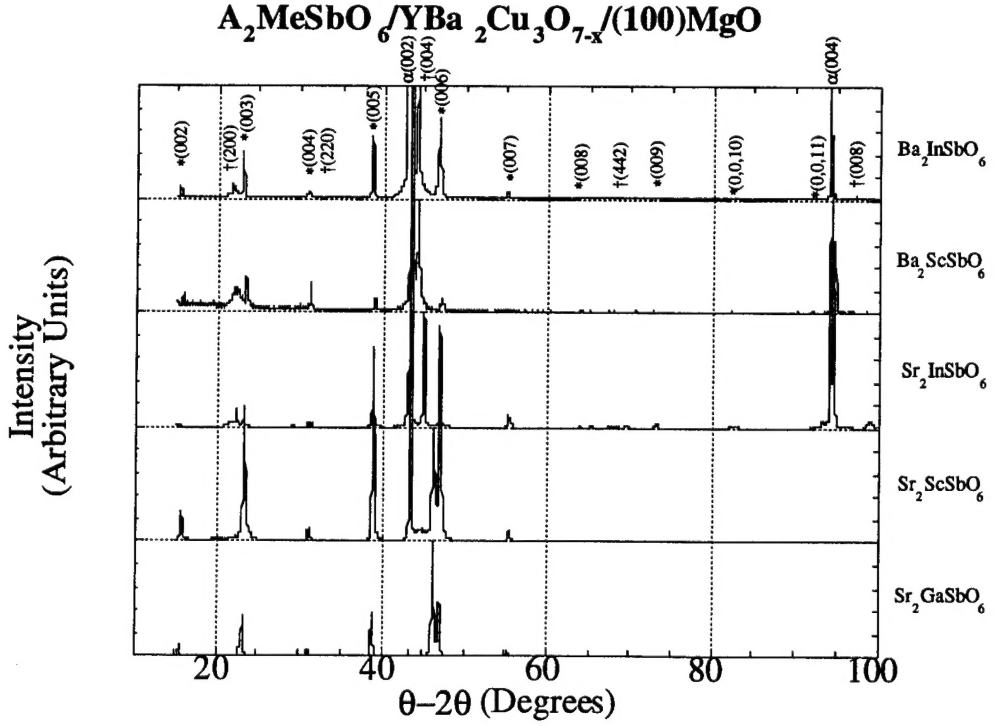

Figure 2. X-ray spectra of A_2MeSbO_6/YBCO/(100)MgO bilayers. The x-ray spectral lines for the YBCO, A_2MeSbO_6 films and MgO substrate are labeled using *, † and α, respectively, and appropriate (*hkℓ*)

indices. The FWHM of the 004 lines of the A_2MeSbO_6 layers ranged from 0.26-0.36 2θ. Cu K_α was used as the radiation source.

TABLE 4. EPITAXIAL RELATIONSHIPS AMONG A_2MeSbO_6, YBCO AND MgO.

Structure	Observed X-ray Reflections		
	MgO	Antimonate	
		Reflection	Intensity
Ba_2InSbO_6/MgO	200	100	very weak
	400	110	very weak
		200	very strong
		400	weak
Ba_2ScSbO_6/MgO	200	200	medium
		220	weak
		400	very strong
		422	very weak
		444	very weak
Sr_2InSbO_6/MgO	200	200	medium
		220	weak
		400	strong
		422	weak
		440	weak

4. SUMMARY

Five compounds in the series A_2MeSbO_6, where A = Ba, Sr and Me = Sc, In and Ga, most of which are ordered perovskites, were successfully prepared by a solid-state reaction from oxide carbonate precursors. Epitaxial growth of thin-film A_2MeSbO_6 on YBCO by pulsed-laser ablation was demonstrated. X-ray diffraction lines for the epitaxial films were quite sharp and the added presence of a high-order reflection attested to the high quality of some of the films. Most of the A_2MeSbO_6 compounds possess dielectric constants smaller than substrate/buffer layers most commonly used, and most have quite acceptable loss tangents for microwave-device applications. The lattice misfit to YBCO is, in most instances, not comparable to $LaAlO_3$. Nevertheless, these compounds overcome many of the shortcomings of $LaAlO_3$, namely: most are cubic or almost cubic, do not undergo a phase transition on cooling, and have very desirable dielectric properties.

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